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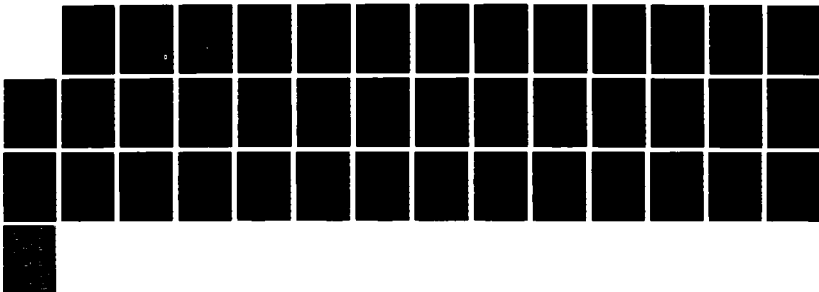
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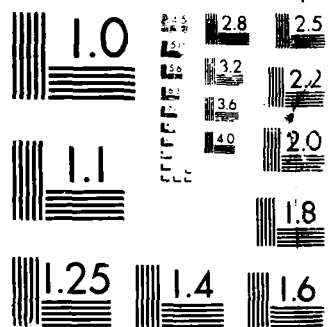
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NONCONTACT PHOTO-THERMAL SENSING

N00014-83-C-0170

FINAL REPORT

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TABLE OF CONTENTS

1. Introduction
2. Research Achievements
 - 2.1 Travelling Thermal Lens (TTL) Spectroscopy for a Particulate-Free Flowing Gas
 - 2.2 Photo-Thermal Modified Backward Mie Scattering from an Aerosol Particle
 - 2.3 Photo-Thermal Radiometry Signal Observed from a Single Aerosol Particle
 - 2.4 Thermal Diffusivity and Temperature Measurements in an Unconfined Hot Gas
 - 2.5 Photo-Thermal Detection of Photochemical Particulate Production
 - 2.6 Novel Optical Method for Ultrasonic Absorption Spectroscopy
 - 2.7 Optical Detection of Photo-Acoustic Pulse Propagation in Silicon Wafers
 - 2.8 Remote Measurement of Properties of Thin Films
 - 2.9 Ultra-Short Acoustic Pulses Generated by Lasers
 - 2.10 Remote Measurement of Absolute Absorption Coefficients
 - 2.11 Remote Detection of Layered Structure
 - 2.12 Noncontact Detection of State of Agglomeration Powders
 - 2.13 Quantitative Measurement of Subsurface Thermal Properties Below an Opaque Film
 - 2.14 Monitoring Amounts of Surface-Adsorbed Molecules in Atmospheric Conditions
 - 2.15 New Optical Techniques to Measure Magnification of a Thin Magnetic Film Deposited on a Substrate
3. References
4. List of Publications Supported in Part by this Contract
5. Invited Papers on This Contract Supported Work Presented in International Conferences
6. Honors Received Partly Due to Contract Supported Work
7. Resume

1. INTRODUCTION

This Final Report summarizes the research efforts and achievements of Contract N00014-83-C-0170, which was first funded in January 17, 1983, and was renewed in January 1984, 1985 and 1986. The research program was concerned with the development of noncontact photo-thermal sensing techniques useful for chemical and physical identification of distant samples. The goals of our research efforts include the exploration of various chemical and physical applications of noncontact photo-thermal sensing, understanding the basic physics and applications of pulsed photo-thermal and photo-acoustic generation, and the understanding of the capabilities of a typical photo-thermal LIDAR (Light Detection And Ranging).

In the five-year contract period (on a 50%-50% cost-sharing basis), we have worked towards the above goals, and have made significant advances and achievements. The traditional LIDAR relies on the detection of luminescence, Raman scattering, Rayleigh or Mie scattering from distant sample that is excited by one or more pulsed laser beams, and provides spectroscopic information on distant sample if there is significant luminescence or Raman scattering. In contrast, this contract study indicates that a "photo-thermal LIDAR" not only can provide spectroscopic information on distant nonluminescent samples, but also other information on physical properties like layered structure, thin-film thickness, degree of aggregation of particles, and flow velocity and composition of a distant cloud. In addition, the basic physics and significant applications of pulsed photo-thermal generations and associated effects like the photo-acoustic (or opto-acoustic) effect are studied. These achievements are summarized in this report and extensively published.

2. RESEARCH ACHIEVEMENTS

Summary of significant achievements are given here; details can be found in the publication cited here. Also, a complete list of all publications supported in part by this contract work is given in Section 3.

Part A: Non-Contact Photo-Thermal Sensing Achievements

For Aerosols/Particulates in Gases

2.1 Travelling Thermal Lens (TTL) Spectroscopy for a Particulate-Free Flowing Gas

Optical methods are important for the studies of flows as they create practically no perturbation, and can easily be applied to hostile environments to provide information on flow patterns and composition. Most of previous methods rely on Mie scattering from particulates seeded in the flow; these include interferometric methods like laser Doppler anemometry,¹ speckle interferometry,² or imaging³ techniques. Coherent Raman scattering techniques⁴ can be used to measure velocity, concentration and temperature profiles in plain unseeded vapors. We have applied⁵ pulsed photo-thermal heating to flows of a plain gas or an aerosol-containing gas to obtain flow and spectroscopic information. This new method complements previous methods since measurements are now possible in unseeded gas with low flow velocities. In an unseeded gas, pulsed laser heating creates a thermal lens which moves with the flow, and can be probed downstream by a continuous-wave (cw) laser.

In our experiment, a tunable Q-switched CO₂ laser beam (pulse duration 1 μ s, energy 0.1 mJ) is focused onto the flow. The absorption of the infrared light by the vapor leads to the formation of a moving refractive index distribution, *i.e.*, a TTL, which moves with the flow and is probed by a cw laser beam downstream as a beam deflection signal. The cw probe beam from a diode or a HeNe laser is directed perpendicular to the pump beam for good spatial resolution, which is determined by both focal diameters. In this configuration, the thermal lens corresponds to a thin cylindrical lens,^{6,7} in contrast to most conventional thermal lensing spectroscopy using coaxial probe and pump beams corresponding to a thick spherical lens. The probe beam deflection is detected using a bicell detector which effectively eliminates noise due to probe intensity fluctuations.

The TTL deflection angle $\phi(t)$ as a function of time t is calculated⁵ as

$$\phi(t) = -\frac{1}{n} \frac{\partial n}{\partial T} \frac{8\alpha DE(vt - a)}{\sqrt{2\pi} \lambda (w^2 + 8Dt)^{3/2}} \exp\left[-\frac{2(vt - a)^2}{w^2 + 8Dt}\right] \quad (1)$$

where n is the refractive index, T is the temperature, E is the pump laser pulse energy, w is the pump laser focal radius, α is the absorption coefficient of the medium, v is the flow velocity, a is the pump and probe beam separation, D is the thermal diffusivity, and λ is the thermal conductivity of the medium. Under static (zero flow) conditions, $(vt - a)$ is simply $(-a)$, which is typically set at $\pm w/2$ to maximize signal in a static probe beam deflection experiment. This may be compared with the results of Refs. 6 and 8, where static thermal lensing and collinear pump and probe beams are used, respectively. Experimental curves obtained for different beam separations show that the signal is more delayed, weakened and broadened with increased beam separation due to thermal diffusion; besides the flow velocity, thermal diffusivity D can be derived which provides temperature information.⁸ The determination of the flow velocity is straightforward by measuring the zero-crossings of the signals. The width of the signals is determined both by thermal diffusion and the flow velocity, and this can be used to calculate instantaneous velocities, *e.g.*, in turbulent flows. Complications arise in highly turbulent flows, since the strong density gradients can cause additional probe beam deflections.

2.2 Photo-Thermal Modified Backward Mie Scattering from an Aerosol Particle

We have demonstrated a new single-ended technique for obtaining the flow velocity and spectroscopic information of aerosol particles by photo-thermal modified backward Mie scattering. These experiments were performed on a nitrogen flow seeded with water aerosols from an ultrasonic nebulizer. Pulsed laser heating causes the water aerosols to partially evaporate which is detected by transient Mie scattering of the probe beam after a transit time. For these experiments, probe and pump beams were at a small angle to each other to increase the interaction volume. At higher power levels (above 2 J/cm^2), almost complete particle evaporation is observed in agreement with previous investigations.⁹

The flow velocity of the aerosols is obtained either by analyzing the time delay of the deflection signal as a function of beam separation a , or by measuring the deflection signal width which is determined by the flow velocity and the geometrical width of the heated region. This technique may also be useful to study turbulence by observing signal broadening. It should be noted that spatial resolution is in general kept lower for these observations to reduce the effect of particle number fluctuations in the probed volume. As this technique is *single-ended* with the lasers and detection system on one side of the flow, this method is especially attractive for remote sensing applications.

2.3 Photo-Thermal Radiometry Signal Observed from a Single Aerosol Particle

A direct way to measure photo-thermal heating of particles is to measure the increased thermal radiation^{10,11} by using a suitable IR broadband detector. These experiments were carried out on a stream of monodispersed ethylene glycol droplets from a droplet generator consisting of a piezoelectric tube and a nozzle. (droplet diameter 40-60 μm). The stream of droplets is crossed by a cw tunable CO_2 laser at 10.2 μm so that the particles are heated during their transit time. Infrared radiation from the particles is monitored by a broadband HgCdTe detector (New England Research, 1 MHz bandwidth) with a MgF_2 window to suppress the stray CO_2 laser scattered light. The IR emission detected is, therefore, limited mainly to the 5-9 μm spectral region, as longer wavelengths are suppressed by the entrance window. Absolute temperature measurements can be obtained by calibrating the apparatus. This is done by heating the nozzle to a known temperature, and measuring the IR radiation of the uniformly heated droplets produced. The arrangement is sensitive enough to detect thermal emission from a single moving particle. Experimental results have been obtained as a function of distance between the crosspoint with the CO_2 laser and the observation region. For these measurements, the detector was shifted in equidistant steps and the IR signal observed at a delay time corresponding to the time-of-flight of the particles from the excitation to the observation region. As the droplets are slowed down in the standing air surrounding them, the time delay between successive

measurements increases. The width of the signals is determined by the transit time of the droplets through the observation region. Therefore particle deceleration also causes the broadening of the signals observed. By analyzing the signal delay and the width, the velocity curve of the particles can be reconstructed. Apart from signal delay and broadening, a signal decrease is also observed for increased separation. This is caused by nonuniform particle heating, which yields a stronger initial thermal emission intensity due to the T^4 dependence of blackbody radiation. The temperature equilibration time for a particle of radius r is characterized by $t_e = 4r^2/D$ where D is the thermal diffusivity of the particle medium. For $60\text{ }\mu\text{m}$ ethylene glycol droplets, this yields $t_e = 4\text{ ms}$ in good agreement with the relaxation time observed in the experiment. From the emission intensity at long times, information on the absolute temperature can be derived, which is a direct measure of particle absorption. This can therefore provide single particle absorption spectra with absolute calibration.

2.4 Thermal Diffusivity and Temperature Measurements in an Unconfined Hot Gas

We have made a first experimental study and analysis of the evolution of a transient thermal refractive index gradient (RIG) produced by a pump laser pulse and detected by a spatially separated probe beam. We show that thermal diffusivity D or gas temperature can be obtained by a photo-thermal probe beam deflection (PTPD) technique, unlike most previous PTPD investigations which are mainly spectroscopic measurements. A notable exception is the work of Murphy and Aamodt¹² who used a "mirage" deflection effect near solid target excited by a modulated thermal source to determine the thermal diffusivity of the gas in contact with the target; their method is not totally noncontact because the insertion of the suitable target into the gas is needed.

Our present work demonstrates a new method for noncontact monitoring of temperature or material composition that affects D . The present experiment is designed to measure the thermal RIG in an unconfined hot gas to mimic an open flame. A pulsed CO_2 laser beam is used to produce a transient thermal refractive index gradient in a

nitrogen gas doped with trace amounts of absorbing Freon 12 at temperatures from 25 to 425°C. The diffusion of this gradient is probed by a continuous helium-neon laser beam parallel to but displaced from the pulsed beam. The observed deflection signal agrees well with theory, and thermal diffusivity or gas temperature can be derived from the signal. Such a nonintrusive method should thus be valuable for measurements *in situ* in open media like flames or other hostile environments. Details of this work have been published recently.⁸

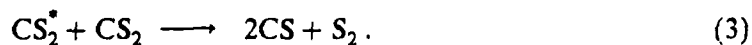
2.5 Photo-Thermal Detection of Photochemical Particulate Production

We have observed a new application of PTPD, namely, the monitoring of photo-chemical particulate production¹³ process. Although the phenomenon of photo-chemical particulation (PCP), *i.e.*, nucleation and growth of particles due to a chemical reaction induced by light (also called "Laser-Snow" for laser-generated particles), is known in many vapor systems, all previous monitoring of PCP has been performed by Mie scattering, which is not sensitive when the particles are small in numbers or in size. In particular, Mie scattering is not well suited for detecting the onset of PCP with high time resolution. The present technique of PTPD detects the heat of condensation released due to the PCP process. Although heat of condensation is expected to be an important parameter affecting the rate of particle growth in an aggregation process, the present work appears to be the first experimental monitoring of this heat *in situ*. Thus, PTPD not only should provide better understanding of PCP, but also nucleation and particle growth in general.

The experimental arrangement for PTPD monitoring of PCP is performed in a CS₂ vapor. The excitation N₂ laser beam first passes through a 0.8 cm aperture. Its pulse energy is less than 0.1 mJ and full width at a half maximum is 8 ns. It is focused by a lens of 18 cm focal length into the center of the quartz cell to a horizontal width of 2 mm and a vertical Gaussian diameter of 350 μ m. The probe beam is a 0.5 mW HeNe laser beam focused by a lens of 10 cm focal length into the center of the cell

perpendicular to the excitation beam. The focused beam waist of the HeNe laser is 40 μm . In this geometry, the HeNe laser only probes the heat diffusion perpendicular to the plane defined by the two laser beams, as heat diffusion in the direction of the probe beam is compensated by an increased interaction length. Usually, the HeNe focus is adjusted for maximum PTPD signal. This maximum occurs in an intensity wing of the excitation beam and this also allows us to determine the pump beam width. The deflection signal decreases in magnitude but remains same in shape as the HeNe beam is moved more towards the center of the N_2 laser beam. The probe beam deflection is monitored by a bicell detector (Silicon Detector Corporation) that is symmetrically located with respect to the HeNe beam. The bicell detector outputs are fed to a differential amplifier (6 MHz bandwidth), and the amplified difference signal is accumulated on a transient recorder (Data Precision Model Data 6000). This differential detection scheme eliminates most of the noise due to probe beam intensity fluctuations.

The dependence of the observed signals on the vertical displacement of the centers of the N_2 beam and the HeNe beam has been investigated; if the displacement is much larger than the N_2 beam size, we mainly observe the photo-acoustic refractive index profile, while the diffusive thermal wave can only be detected near the source. Here, we only describe the thermal refractive index profile detected by overlapping beams (*i.e.*, probe beam at the $1/e^2$ -waist of the pump beam) for maximized signal amplitude. At CS_2 pressures below ~ 10 Torr, the PTPD signal is simple, with a fast rise and a slow decay. However, at CS_2 pressures above 10 Torr, the PTPD signal becomes more complicated and a second slow rise develops at higher pressures. This additional signal cannot be explained by collisional relaxation which becomes faster at higher CS_2 pressures. The slow heat release can be explained by the photo-chemical reactions induced by the UV laser, for example,¹³



Reaction (3) may already occur at pressures below ~ 10 Torr; however, in this pressure region, the concentrations of the products are below the condensation thresholds. Above about 10 Torr the critical concentration is reached where nucleation starts, *i.e.*, some clusters are above the critical size which is needed for growth. Experimentally, we found that particulates can be observed by light scattering after a few hundred N_2 laser shots at pressures above 20 Torr. Below 10 Torr no particulates can be detected after ten thousand N_2 laser shots. These considerations suggest that the slow PTPD signal at higher CS_2 pressure is due to a delayed heat release caused by the aggregation of particles in the irradiated region.

We have collected some particulates for analysis by ESCA (electron spectroscopy for chemical analysis). This yields a composition of C and S in the ratio 1:1. This ESCA result indicates that under our experimental conditions, only CS condenses into particulates and sulphur molecules stay in the gas phase probably as S_8 unit. SEM (scanning electron microscopy) results furthermore indicate a spherical geometry with diameters around $0.2\ \mu m$. Details of this work is published in Ref. 14.

2.6 Novel Optical Method for Ultrasonic Absorption Spectroscopy

The absorption spectrum of high frequency sound (frequency f much larger than 1 MHz) in gases has traditionally been very difficult to measure. This is because of the lack of a controllable high-frequency source with good acoustic coupling to the gas, and lack of a sensitive high bandwidth detection method. Thus, classical methods using ultrasonic transducers are usually limited to frequencies less than 1 MHz. The propagation of high-frequency waves (*e.g.*, with f approaching a molecular relaxation frequency, or wavelength approaching a collisional mean-free-path) remains to be an open question for many gases. We have made a first demonstration of an all optical Fourier-multiplexed method to measure ultrasonic absorption spectra of gases and mixtures near normal temperature and pressure conditions at frequencies up to tens of MHz. Our technique is new in two respects: (1) It is all optical, and hence is noncontact

and applicable for corrosive or inaccessible samples. (2) It is a frequency-multiplexed technique relying on pulse-broadening measurement and Fast Fourier Transform (FFT) analysis, and hence is fast and applicable for real-time measurements under rapidly changing conditions.

In our experimental demonstration, the gas being studied is contained in a temperature controlled quartz cell. A pulsed N_2 laser beam (1 mJ energy and 8 nsec duration) is directed to an absorption target (highly polished Si wafer) in the cell to produce a narrow acoustic pulse. The laser spot size at the target is 2 mm, and no material change at the target occurs. A continuous HeNe laser beam is carefully focused into the cell such that it is parallel to the target surface and the focus is vertically above the irradiated spot of the target. The acoustic pulse produced at the irradiated target propagates through a distance x before it arrives at the probe laser focus causing a transient deflection of the probe beam. The distance x can be finely adjusted by a micrometer. The probe deflection is converted into an intensity variation by using a small photodiode suitably positioned at the wing of the transmitted probe beam. The amplified signal is displayed on a Tektronix 7854 scope and accumulated on a Data Precision Model 6000 transient recorder with FFT capability. Observed signals for a gas mixture of $CO_2 + 20 \text{ Torr } H_2O$ at 1 atm pressure and $22^\circ C$ clearly shows the pulse broadening with propagation distance and FFT provides ultrasonic absorption spectra. More details of this novel optical technique for ultrasonic absorption spectroscopy have been published.¹⁵

Part B: Non-Contact Photo-Thermal Sensing Achievements for Thin Films

Condensed Matter, Layered Structure, and Powders

2.7 Optical Detection of Photo-Acoustic Pulse Propagation in Silicon Wafers

We have developed a beam deflection method for optical monitoring of nanosecond photo-acoustic pulses in silicon wafers for noncontact and nondestructive evaluation of material properties. Nanosecond acoustic pulses are produced in thin

silicon wafers by the absorption of weak nitrogen laser pulses causing no sample ablation. A HeNe probe laser is tightly focused onto the surface of the same or the opposite side of the wafer. Deflections of this probe beam are monitored by a photodiode located in the Gaussian wing of the reflected, diverging beam. The transient deflection signal can be observed on a Tektronix Model 7104 oscilloscope in a single shot at nanosecond time resolution.

The absorption of the pump laser pulse causes a surface distortion, which is the source of longitudinal and shear wave pulses propagating into the sample. This can be detected on the same or opposite side of the sample with the probe beam focus located in the wing of the excited spot. As this technique measures the surface gradient, the signal changes sign when shifting the probe beam focus across the excited spot and vanishes when it is exactly centered. In agreement with experiments measuring surface height distortions, we observe an initial negative gradient caused by the longitudinal wave followed by the shear arrival at a later time causing a positive gradient. Overlayed on this is a weak "oscillation" which we interpret as multiple echoes of a highly collimated, longitudinal pulse in the thin wafer.

If the probed spot is shifted from the excitation position, the propagation of Lamb waves in the thin wafer can be observed. In our experimental conditions, we mainly excite low-order Lamb waves. The lowest order antisymmetric a_0 wave is characterized by a strong dispersion, approaching zero velocity at low frequencies and the Rayleigh wave velocity in the high frequency limit. This is the reason why a "frequency-chirped" surface wave pulse is observed. Here, surface wave signals observed at a distance from the excited spot start at a well-defined delay time proportional to the beam separation, and continue afterwards according to the different arrival times of the different modes excited by the short laser pulse. If the excited and probed spot are overlapping in the center of a round wafer, multiple reflections from the circumference

can be observed. As the corresponding signal depends sensitively on the alignment of the excited spot, this signal can be used for accurate positioning of the wafer.

This work shows that the probe beam deflection technique is a fast and simple method for measuring acoustic pulse propagation in solids.^{16,17} It does not require the elaborated mechanisms needed in interferometric detection schemes while offering similar sensitivity. In addition, it does not require high surface quality of the medium under investigation. Due to its noncontact nature, it is applicable to hostile environments. Extensions of probe-beam deflection technique to nonacoustic events like monitoring moisture adsorption on a surface or measuring magnetostriction of a thin film have also been recently demonstrated and mentioned in Sections 2.14 and 2.15.

2.8 Remote Measurement of Properties of Thin Films

We have developed theoretical analysis as well as experimentation for single-ended, noncontact and nondestructive measurements of various types of thin-film or layered samples, including metal films, polymer films, and paper (*e.g.*, that of the paper currency of the United States). Details of the experimental results are published.^{18,19}

Our experimental arrangement utilized a short pulsed N_2 laser beam to rapidly heat up the sample surface (*e.g.*, a few °C). No sample damage occurs, since the laser pulse (of energy less than 1 mJ and of 8 nsec duration) is slightly focused to a spot size of about 4 mm diameter at the sample. The back-emitted IR radiation due to the transient photo-thermal excitation is detected by a fast HgCdTe detector, and the signal is accumulated on a Tektronix 7854 scope with a 7D20 plug-in. The signals for thin film samples are characterized by the following features. There is an initial fast decrease of the signal with time, and a final "flattening" of the signal after the deposited heat at the sample surface is uniformly distributed through its thickness. The initial fast decrease is observed to be independent of the sample thickness, but depends on the optical absorption lengths and the thermal diffusivity D of the sample. IN contrast, the final

flattening of the signal is found to depend only on the sample thickness and D . Our theoretical fit of the observed signal for the 12.5 micron thickness stainless steel shim at long times produces a value of $D = 0.036 \text{ cm}^2/\text{sec}$, in agreement with other values measured for Type 302 stainless steel. Similar PPTR measurement on a teflon film of 89 micron thickness yields a value of $D = 0.00084 \text{ cm}^2/\text{sec}$, in agreement with the accepted value of $0.00088 \text{ cm}^2/\text{sec}$. It is evident that our single-ended noncontact PPTR technique provides quantitative values of D ; also, any physical or chemical modifications affecting D of the sample can be remotely monitored.

2.9 Ultra-Short Acoustic Pulses Generated by Lasers

We have made the first demonstration²⁰ of the nondestructive photo-acoustic (PA) generation and detection of 1 nsec acoustic pulses in condensed matter. These pulses are at least an order of magnitude narrower than those previously reported. The ultra-short PA pulses here are generated by a high-pressure N_2 laser which produces pulses of 0.5 nsec duration, and detected with thin-film ZnO transducers. The ultra-short PA pulses are shown to be ideally suited for measuring sample thickness l , acoustic velocity c , or acoustic attenuation α in a thin film by observing the multiple echoes. Accuracy in thickness measurements of 1% is demonstrated for the $12 \mu\text{m}$ thick stainless steel films once c is known; such high accuracies for thin films of $10 \mu\text{m}$ thickness were previously impossible with pulsed transducer measurements. We have also shown the measurement of α , which is known to be related to important thin-film properties such as grain size distribution and crystallinity. The measurement of position-dependent ultrasonic absorption by the present short-pulsed PA technique should provide a new imaging tool for characterizing thin films and locating defects.

We have also demonstrated another application of short PA pulse generation for detecting small elasticity anisotropy in solids. This is demonstrated for extruded bars of aluminum alloys (6061-T6), where we measured the longitudinal velocity $V(\theta)$ as a function of the propagation direction at an angle θ with respect to the extrusion

direction. We observed that $V(45^\circ)$ is the largest, namely 1.8% larger than $V(0^\circ)$ while $V(90^\circ)$ is 0.49% larger than $V(0^\circ)$. These data illustrates that PA pulse generation is a useful and quick technique for measuring and understanding anisotropy in solids.

Details of this work have been published.²¹

2.10 Remote Measurement of Absolute Absorption Coefficients

We have discovered that PPTR can also provide the *absolute* absorption coefficients of strongly absorbing materials.²² This is quite remarkable, because absolute absorption coefficient is very difficult to measure in a single-ended manner by all previously known techniques. The present demonstration of single-ended absolute absorption coefficient measurement suggests that remote trace-detection or chemical identification is possible even with a single nontunable excitation pulsed laser.

The qualitative explanation of the measurement of absolute absorption coefficient b is as follows. If b is large (small), the excitation laser is absorbed over a small (large) depth into the sample, thus setting up a large (small) thermal gradient at the sample surface, causing large (small) thermal diffusion and quick (slow) surface cooling; hence, the PPTR signal decays quickly (slowly). This qualitative description can be quantified, and we show that by fitting the PPTR signal shape to a theoretical form, b can be derived if thermal diffusivity of the sample is known.

We have demonstrated the derivation of b for several types of solid or liquid samples. An example is the case of an opaque black rubber sample of thickness about 5 mm. We first measured the PPTR signal excited by the flashlamp pumped dye laser at 590 nm. Then, the fitting of this observed signal shape to a theoretical form²² is done to produce a value of the absorption length b^{-1} being $1.4 \mu\text{m}$ at 590 nm. To verify this, we use a "Microtome" sectioning system to produce a $3 \mu\text{m}$ slice of the sample; the 590 nm transmission through this rubber slice on a quartz substrate is about 10%, indicating an absorption length b^{-1} of $1.3 \mu\text{m}$. This experiment clearly shows for the first time that the PPTR technique is useful for the remote measurement of b , providing

a value in agreement with the conventional Microtome sectioning and transmission measurement, which is double-ended, time-consuming and destructive.

2.11 Remote Detection of Layered Structure

We have discovered that PPTR may be used for remote sensing of layered structures, which produce characteristic features on the time-evolution of the PPTR signal. To show this, we put a thin transparent polyester coating (thickness about 45 μm) onto the black rubber sample mentioned above. The observed PPTR signal for the coated sample shows an initial fast decay (due to the IR emitted from the black rubber surface and transmitted through the coating) and subsequently a peaked signal at late times (due to the heat diffusion from the rubber surface). This permits the derivation of the coating thickness d from the thermal diffusivity D of the coating material ($D = 1 \times 10^{-3} \text{ cm}^2/\text{sec}$) and the time t_p of the diffusion peak of the PPTR signal, ($t_p = 8 \times 10^{-3} \text{ sec}$), using a one-dimensional thermal diffusion formula:

$$d = (wDt_p)^{1/2} = 40 \mu\text{m}.$$

This compares very well with a direct measurement of the coating thickness of 45 μm , showing the capability of single-ended measurement of thickness or thermal properties of a transport overlayer by PPTR.²²

2.12 Noncontact Detection of State of Agglomeration of Powders

We have found that PPTR can be used for novel monitoring of the state of the agglomeration of powder particles, *i.e.*, detecting whether the particles are loosely piled with little interparticle bonding, or are aggregated and consolidated with strong interparticle bonding. This technique may have important applications, for example, monitoring dangerous or explosive powdered chemicals at a distance, or sensing the mechanical "strength" of a sandy or clay surface to see whether it is strong enough to support the landing of people or equipment. The PPTR signal observed for a sample of loosely piled carbon-loaded epoxy powder (of mean diameter of about 10 μm) exhibits

a very rapid initial decay over an initial fast decay due to *intraparticle* heat diffusion; this gives a particle diameter equal to $(2Dt_1)^{1/2} \approx 5 \mu\text{m}$, with $D = 1 \times 10^{-3} \text{ cm}^2/\text{sec}$ being the thermal diffusivity of the epoxy material. This is in reasonable agreement with the known particle size. After the initial period t_1 , the IR signal now decays by *interparticle* heat transfer, which is much slower due to the poor thermal contact between the loosely piled particles. However, when the powders are compressed together, we observed that the slowly decaying part of the IR signal virtually disappeared and the IR signal now has only the fast decay component since the slow interparticle heat transfer is significantly reduced. We believe that this is the first observation of noncontact aggregation monitoring.^{22,23}

2.13 Quantitative Measurement of Subsurface Thermal Properties Below an Opaque Film

We have demonstrated the quantitative use of pulsed photo-thermal radiometry for measuring important physical properties between a film and a substrate: we quantify the subsurface air gap thickness below an opaque film, as well as measure the contact resistance between a film or coating and a substrate. This new technique for contact thermal resistance measurement has many important applications, for example, to detect the conditions of a film applied onto a surface, its degree of adhesion, aging and peel-off potential.

As an example to demonstrate the use of PPTS for subsurface thermal quantification, we measure the thermal contact resistance at the interface of a smooth polymer film and a polished metal substrate. This method relies on the heating of the film surface by a short light pulse and detecting the subsequent infrared thermal radiation from the surface. An analytical solution to the heat diffusion equation shows that in a suitable delayed time interval, the infrared signal decays exponentially in time with a time constant related to the thermal contact resistance of the interface. By changing gases in the interface at constant pressures, we are able to separate out the thermal conductance into two components: that due to solid contacts and that due to

gas conduction. The thermal conductance due to gas conduction in the interface is proportional to the thermal conductivity of the gas found in continuum fluid theory, except for He which is more than 30% lower. We believe that the discrepancy in He is partly due to the fact that the mean-free-path of He gas molecules is comparable to the mean width of the gaps in the interface and the classical continuum fluid theory for heat conduction would no longer hold.²⁴

We have also recently shown that PPTS is useful as a nondestructive and noncontact technique to quantify the thermal conductivity of a subsurface epoxy bond underneath an opaque surface. The degree of curing of the epoxy can also be monitored.²⁵

2.14 Monitoring Amounts of Surface-Adsorbed Molecules in Atmospheric Conditions

We have show that trace amounts of adsorbed molecules or agents on a surface can be detected in atmospheric conditions by pulsed photo-thermal probe-beam deflection. This relies on the use of a pulsed laser to desorb the adsorbed molecules, and a probe laser to detect the transient refractive index gradient caused by the desorption. This work can be used to quantify minute amounts of adsorbed molecules on a surface.²⁶

2.15 New Optical Techniques to Measure Magnification of a Thin Magnetic Film Deposited on a Substrate

We have developed a sensitive tool for the measurement of the magnetostriction constant of a thin soft-magnetic film on a substrate. To our best knowledge, our measurement sensitivity is more than an order of magnitude better than prior art, and is adequate for measuring magnetostriction coefficient smaller than 10^{-7} in magnitude for a film as thin as 30 nm on a glass wafer of thickness 200 μm . To make a measurement, the sample wafer in the form of a rectangle is clamped near an edge, and a rotating magnetic field of constant amplitude (sufficient to saturate the film) is applied to the film in its plane; this rotating field causes the film to expand/contract in the direction of magnetization for positive/negative magnetostriction coefficient, and results

in a periodic "warping" of the sample at a frequency that is twice the rotation frequency of the field. A continuous helium-neon laser beam suitably incident onto the sample is used to sense this small periodic warpage; the reflected laser beam is incident onto a position sensor, whose output is normalized to the laser beam intensity (to reduce intensity fluctuation effects) and then fed to a phase-sensitive lock-in amplifier that is locked to twice the field rotation frequency. This sample vibration frequency is selected at a value where the vibration-noise spectrum is relatively quiet, and below the first mechanical resonance frequency of the sample-holder assembly to avoid complications in the signal analysis. To provide long-term reproducibility, a "deflection calibration" signal can be produced by translating the position sensor by small amounts (using a piezoelectric transducer) at a different frequency and lock-in detection of the sensor signal at the fundamental of this translation frequency. This tool has been developed, characterized, and applied to the measurement of the saturated magnetostriction coefficients of ultra-thin soft-magnetic permalloy base films.

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43. A. C. Tam and H. Schroeder, "A New High-Precision Optical Technique to Measure Magnetostriction of a Thin Magnetic Film Deposited on a Substrate," submitted to *IEEE Trans. Magnetics*.

**5. INVITED PAPERS ON THIS CONTRACT-SUPPORTED WORK
PRESENTED IN INTERNATIONAL CONFERENCES**
(None are supported by other agencies)

1. A. C. Tam, "Optoacoustic Spectroscopy and Detection," American Physical Society Meeting, Detroit, March 1984 (Abstract published in *Bull. Am. Phys. Soc.* **29**, 249 (1984)).
2. A. C. Tam and W. P. Leung, "Novel Optical Method for Real-Time Ultrasonic Absorption Spectroscopy," invited paper, CLEO '84 meeting, Anaheim, June 1984 (Paper in CLEO '84 proceedings, 1984, p. 242, published by *Opt. Soc. of Am.*).
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4. A. C. Tam, "High-Sensitivity Photo-Acoustic and Photo-Thermal Detection," invited paper, Fourth International Conference on Photo-Acoustic, Thermal and Related Sciences, Montreal, Canada, August 4-8, 1985.
5. A. C. Tam, "Photo-Acoustic and Photo-Thermal Material Testing," invited paper, 1985 Annual Meeting of the Optical Society of America, Washington, DC, October 14-18, 1985.
6. A. C. Tam, "Photo-Acoustic and Photo-Thermal Spectroscopy," invited paper, International Congress on the Application of Lasers and Electro-Optics, ICALEO 1985, San Francisco, November 11-14, 1985.
7. A. C. Tam, "Review of High-Sensitivity Photo-Thermal Spectroscopy," invited paper, First International Laser Science Conference, Dallas, Texas, November 18-22, 1985.
8. A. C. Tam, "Photothermal Methods for Testing Applications," CLEO '86, San Francisco, June 9-13, 1986.

9. A. C. Tam and H. Sontag, "Characterization of Thin-Film and Layered Structures by Pulsed Photothermal Radiometry," FACSS '86, St. Louis, September 29-October 3, 1986.

6. HONORS RECEIVED PARTLY DUE TO CONTRACT SUPPORTED WORK
(None are supported by other outside agencies)

1. A. C. Tam was elected a Fellow of the American Physical Society on November 3, 1985, partly due to published work supported by this contract.
2. A. C. Tam was appointed by the National Research Council to be a member of the Committee on Recommendations for the U.S. Army Basic Scientific Research. The appointment period was from June 1986 to May 1988, to serve as a consultant to the National Research Council concerning academic proposals submitted.

7. RESUME

A. C. Tam

Professional History

1985-present	Manager, IBM Research Division, Almaden Research Center, San Jose, CA
1979-1985	Research Staff Member, IBM Research Laboratory, San Jose, CA
1978-1979	Member of Technical Staff, Bell Laboratories, Murray Hill, NJ
1977-1978	Assistant Professor, Physics Dept., Columbia University, NYC
1974-1977	Research Associate Columbia Radiation Laboratory, Columbia University, NYC
1972-1974	Research Assistant, Columbia Radiation Laboratory, Columbia University, NYC
1971-1972	Preceptor, Physics Dept., Columbia University, NYC
1970-1971	Faculty Fellow, Physics Dept., Columbia University, NYC
1968-1970	M.Sc. candidate (holding Hong Kong Government scholarship), University of Hong Kong; also part-time teacher of Applied Mathematics, St. Louis College, Hong Kong

Research Activities and Interests

Laser interaction with matter; laser-induced etching, ablation, or deposition; quantum electronics; photo-thermal spectroscopy; photo-acoustic detections of weak absorptions; nonlinear optics; laser-spark generations; optical pumping; photochemistry; atomic and molecular spectroscopy.

Academic Degrees

Ph.D.	Columbia University, 1975. Thesis research: "Stepwise excitation and level crossing spectroscopy of the triplet-D states of Helium-4." Thesis sponsor: W. Happer.
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Fellow, American Physical Society, 1985-present.

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Member, Optical Society of America, 1983-present.

Member, IEEE, 1984-present.

Committee Member, National Research Council, 1986-1988.

Guest Editor, IEEE Trans Ultrasonics, Ferroelectrics, and Frequency Control, September 1986 issue.

Program Chair, International Laser Science III, 1987 (APS Topical Group Meeting)

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11. A. C. Tam, "Application of Pulsed Photothermal Probe-Beam Deflection Measurements in Gases," 3rd International Laser Science Conference, Atlantic City, November 2-5, 1987.
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13. A. C. Tam, "Photothermal Techniques in Materials Characterization Applications," Conference on Progress in Non-Destructive Testing, *Keynote* invited paper, La Jolla, July 31-August 5, 1988.
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